The role of nuclear spins in photoionization: isotope-resolved measurement of the high-n, odd-parity, autoionizing Rydberg states of krypton

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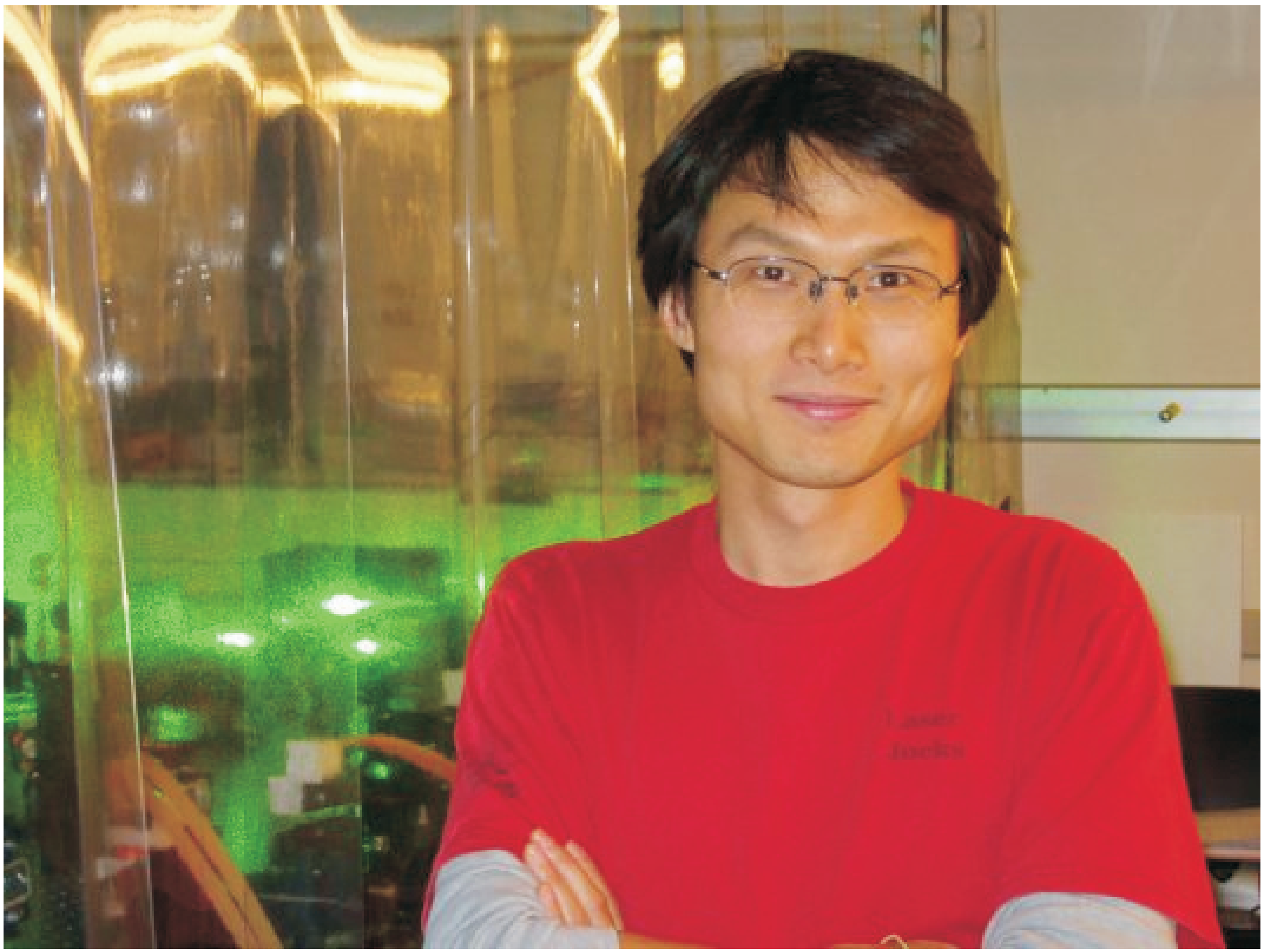


The role of the nuclear spins in photoionization

Current status:

Conservation of nuclear-spin symmetry is assumed (e.g. ortho-para interconversion is forbidden)

Otherwise: Nuclear spins are ignored

^{83}Kr (I=9/2):

$$^{83}\mathrm{Kr}\ (^1S_0, I=9/2, F=9/2) \longrightarrow\ ^{83}\mathrm{Kr}^+\ (^2P_{3/2}, I=9/2, F^+) + e(\ell,s,j)$$

ortho H_2 (I=1):

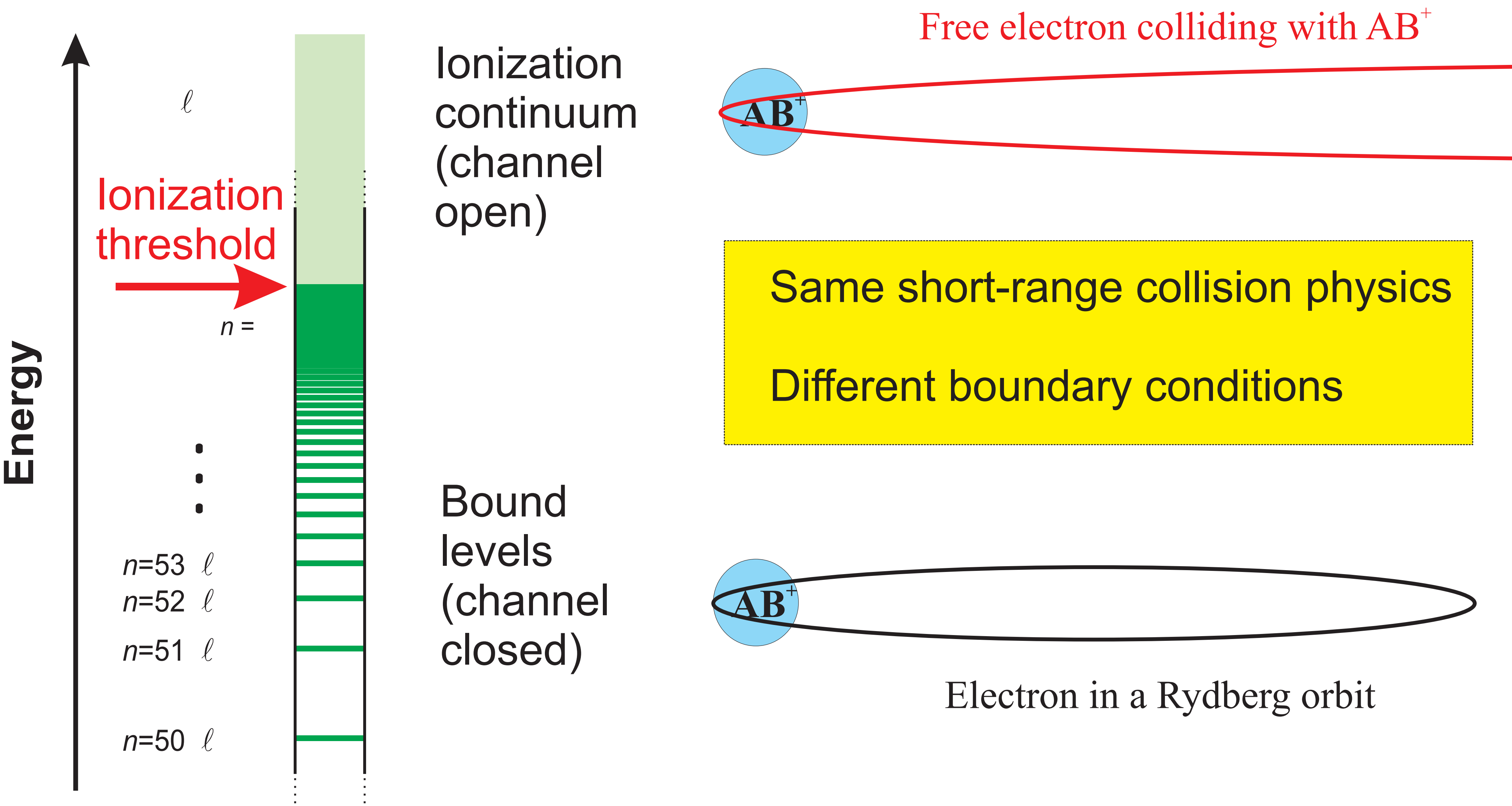
$$H_2(^1\ \ _g, v, S=0, N=J, I=1, F)$$

$$\longrightarrow H_2^+(^2\ \ _g, v^+, N^+, S^+=1/2 J^+, I=1, F^+) + e(\ell,s,j)$$

The hyperfine (dipole-dipole,Fermi-contact) interaction turns on upon ionization. Angular momentum coupling hierarchy changes.

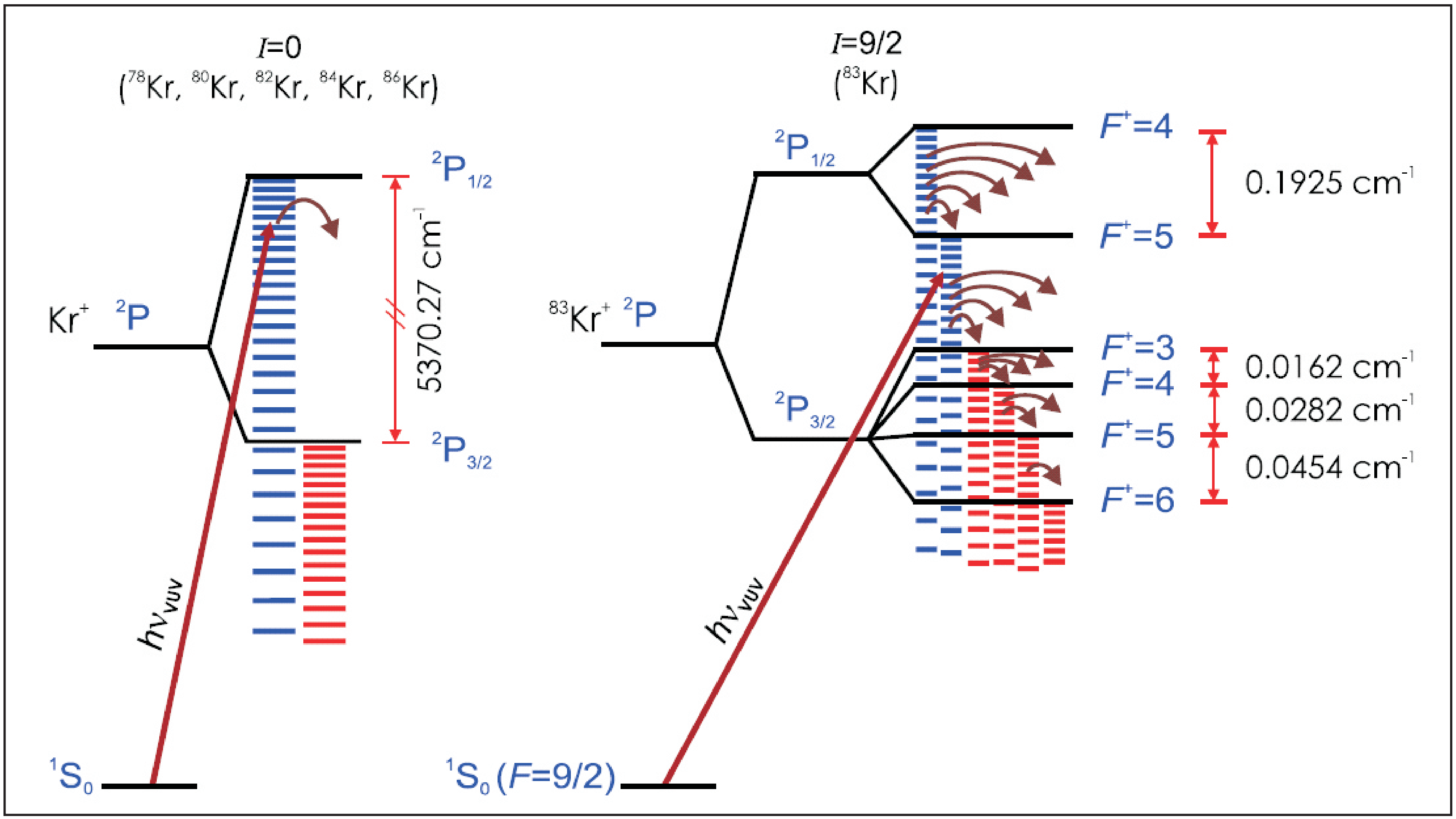
Study photoionization and electron-ion collisions by high-resolution spectroscopy

Ionization channel = Ion in well-defined (v^+, J^+) quantum state + electron of given orbital angular momentum ℓ



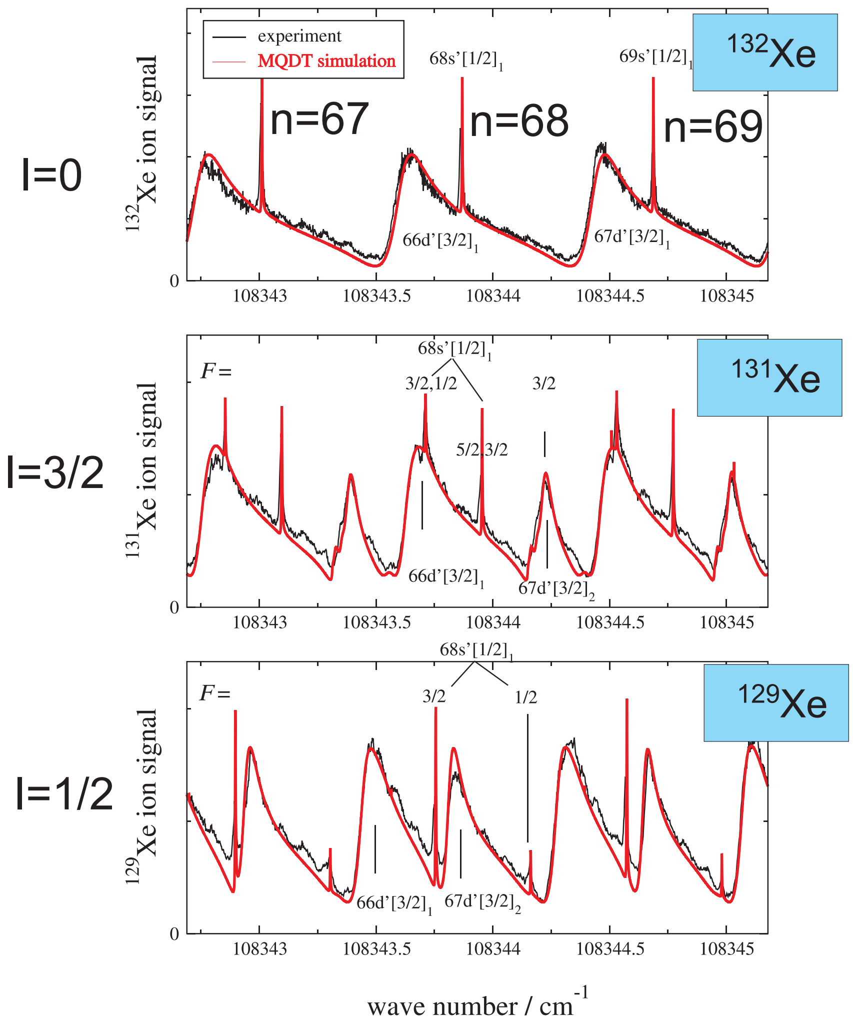
Autoionization dynamics in ^{83}Kr (I=9/2)

Is the photoionization spectrum of ^{83}Kr identical to that of ^{84}Kr?
How does the hyperfine structure of very high Rydberg states look like?
What are the propensities for autoionization?
Is there a pure hyperfine autoionization?
Can one prepare ions in selected F^+ states?
How does one determine the hyperfine structure of the ion from the Rydberg spectrum?



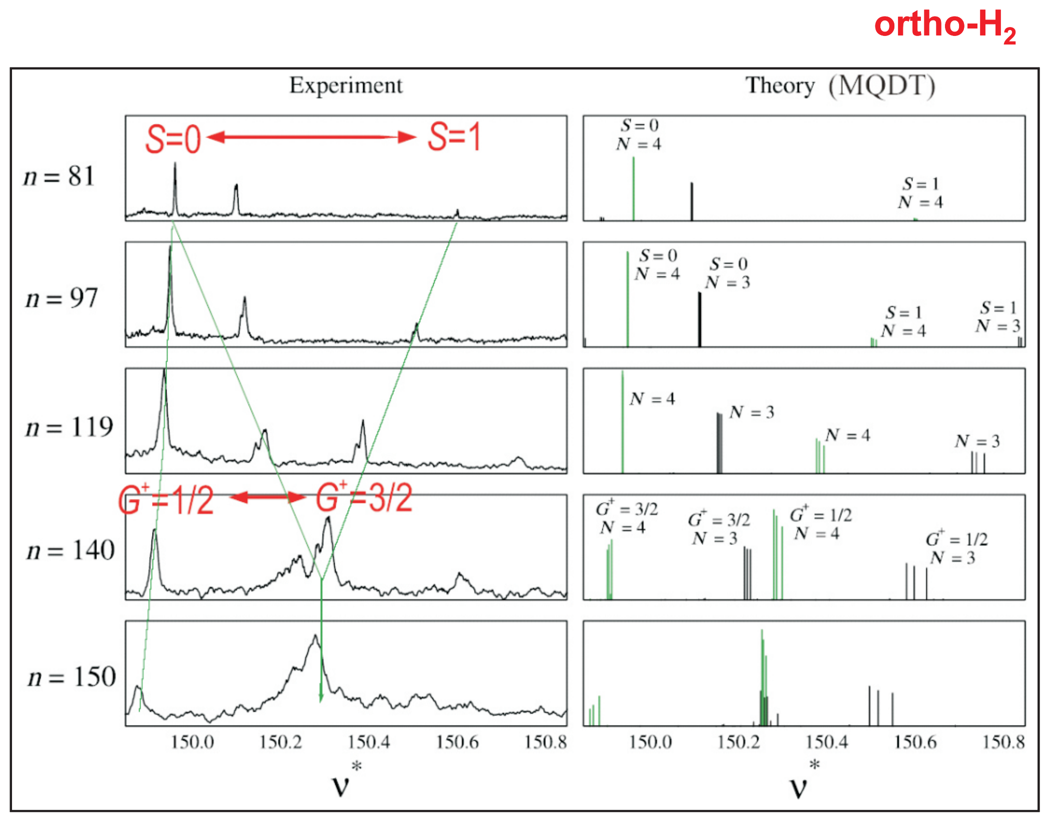
Previous studies

The Beutler-Fano resonances in the photoionization spectrum of atomic xenon



H. J. Wörner et al.,
Phys. Rev. A 71, 052504, 2005

Rotational autoionization dynamics in Rydberg states of ortho H_2

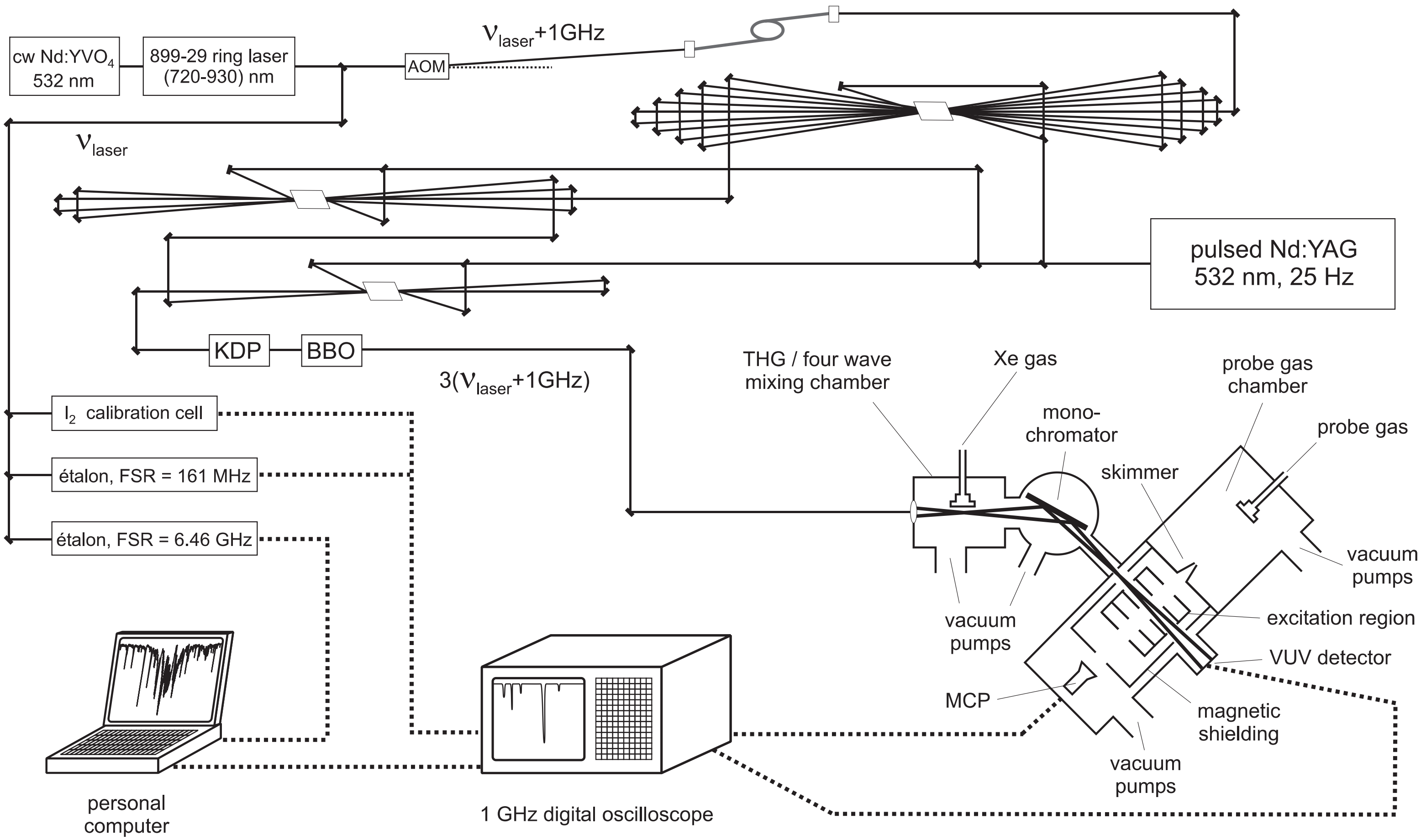


H. J. Wörner et al.,
Phys. Rev. A 75, 062511, 2007.

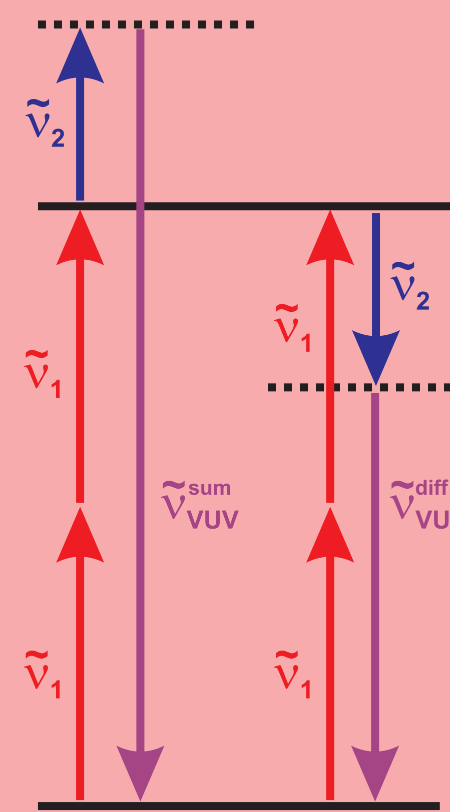
Experiment

Fourier-transform-limited VUV laser with programmable pulse shapes and lengths

Bandwidth: 50 MHz, Tunable range: 92000-118000 cm^{-1}

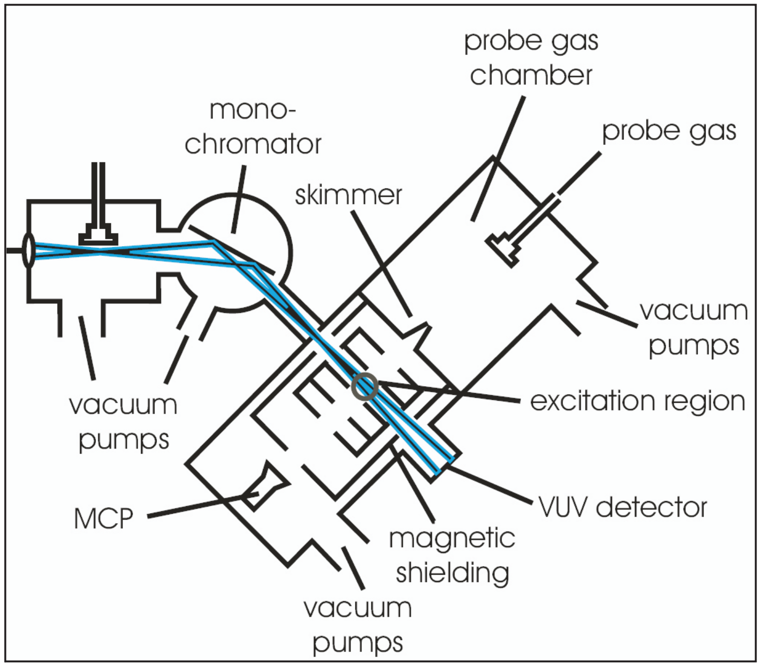


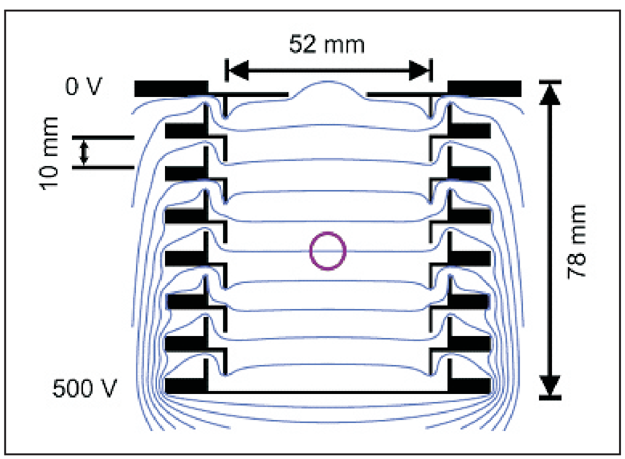
VUV generation by resonance-enhanced four-wave mixing in rare gases (Ar, Kr, Xe)

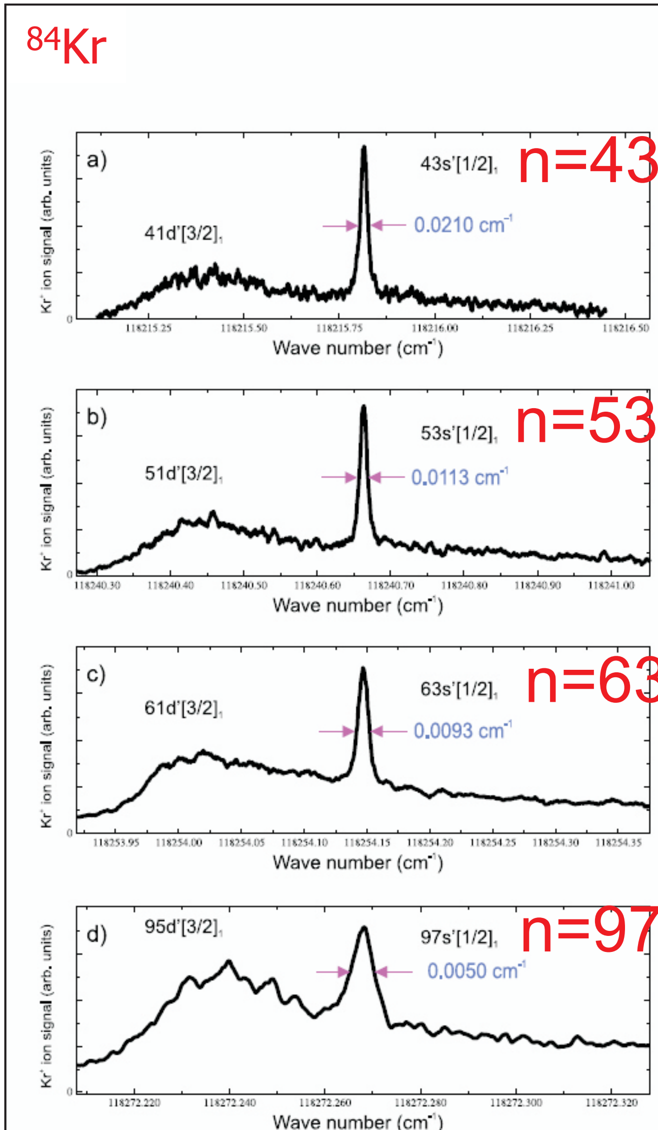


Paul et al., J. Phys. B **38**, 4145 (2005)

Photoionization experiments



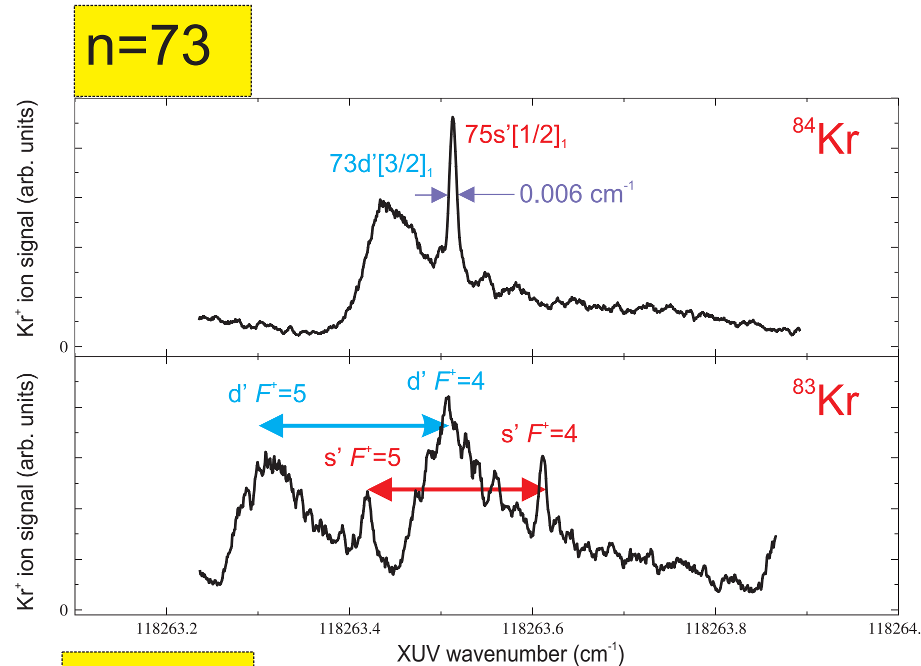


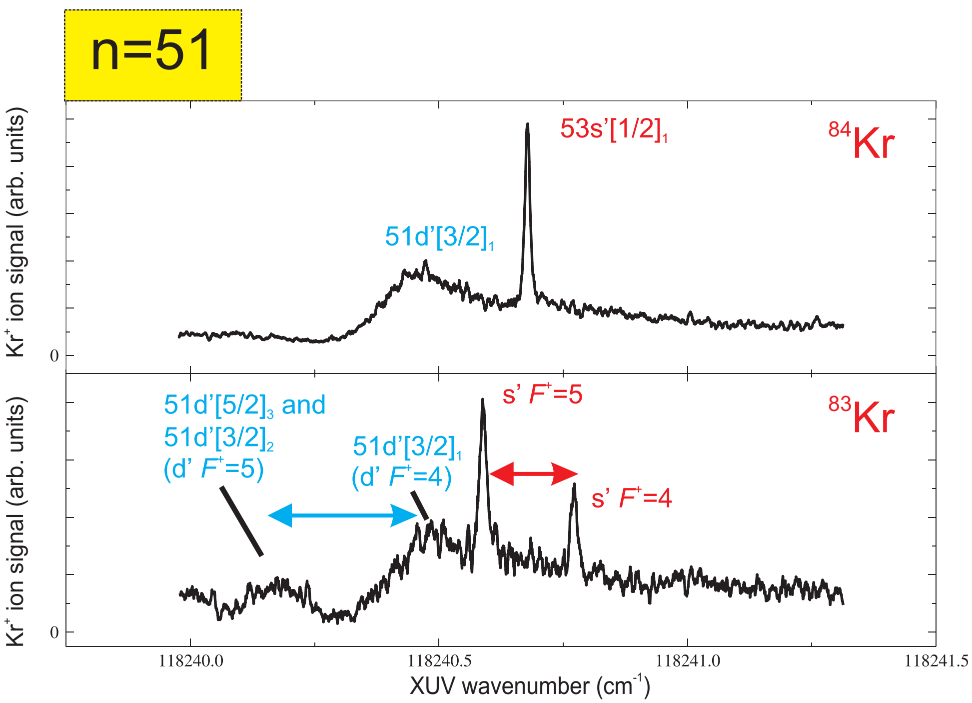


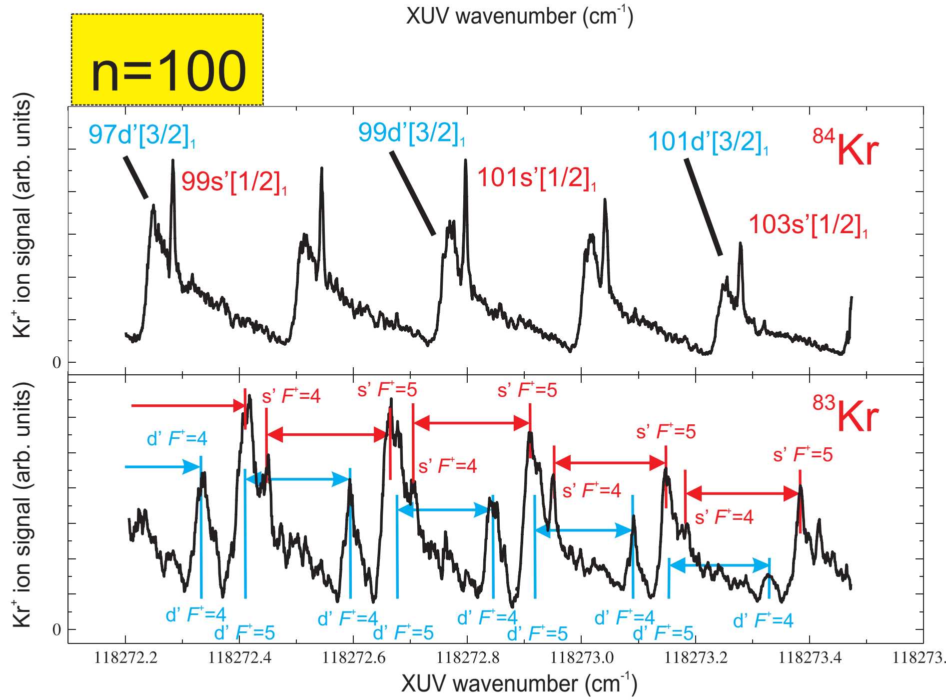
	s′	d′
	2 ns	98 ps
	3.7 ns	200 ps
	6.3 ns	320 ps
	23 ns	1 ns
n=270	0.5 μs	28 ns

Autoionization resonances in krytpon

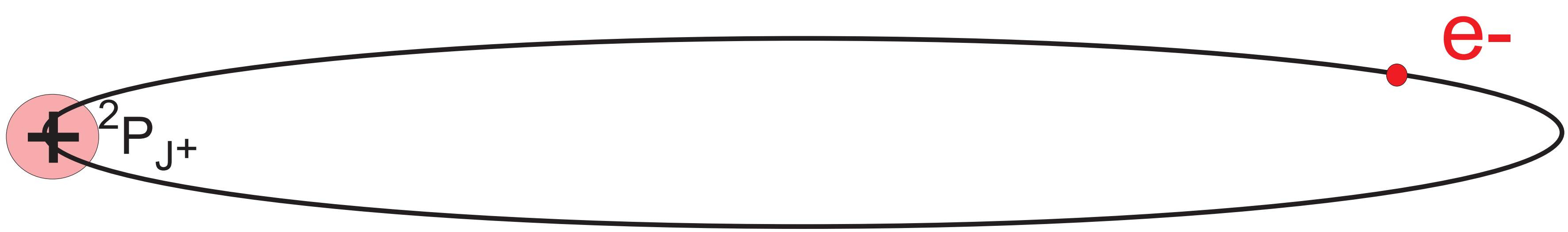
The Beutler-Fano resonances
in the autoionization region of the
photoionization spectrum
of atomic krypton







Multichannel quantum defect theory without hyperfine interaction



K.T. Lu, Phys. Rev. A 4, 579 (1971)   MQDT: Seaton, Fano

Close-coupling region:

Strong interaction between ion core and electron:

Good quantum numbers:
S=0,1, J=0-4
l=0,2

Quantum defects (J=1):
($s\ ^1P_1$)   ($s\ ^3P_1$)
($d\ ^1P_1$)   ($d\ ^3P_1$)   ($d\ ^3D_1$)

Angular momentum frame transformation

Long-range region:

Very weak interaction between ion core and electron:

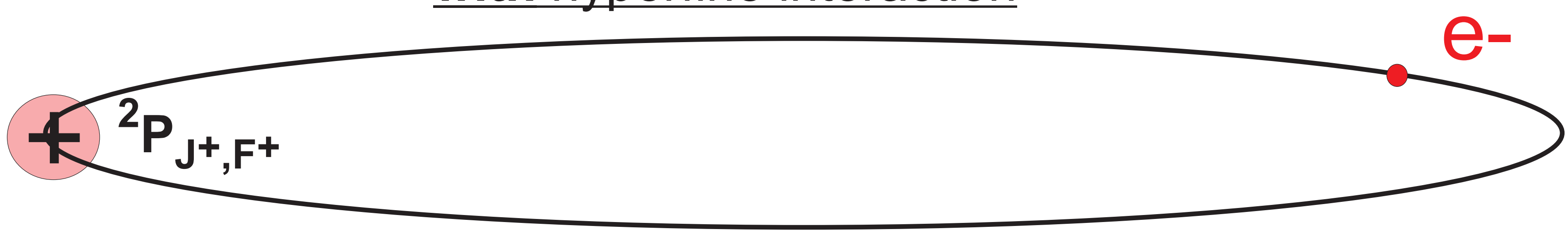
Good quantum numbers J=0-4:

core: S^+=1/2, L^+=1, J^+=1/2,3/2
electron: l=0,2

Quantum defects:
($s\ ^2P_{3/2}[3/2]_1$)   ($s\ ^2P_{1/2}[1/2]_1$)
($d\ ^2P_{3/2}[3/2]_1$)   ($d\ ^2P_{3/2}[1/2]_1$)
($d\ ^2P_{1/2}[3/2]_1$)

For Krypton: M. Aymar, O. Robaux and C. Thomas, JPB 14, 4255 (1981)

Multichannel quantum defect theory **with** hyperfine interaction



Close-coupling region:

Hyperfine interaction negligible compared to core-electron interaction:

Good quantum numbers: F
S=0,1, J=0-4
l=0,2

Quantum defects: unchanged

Angular momentum frame transformation

Long-range region:

Hyperfine interaction larger than core-electron interaction

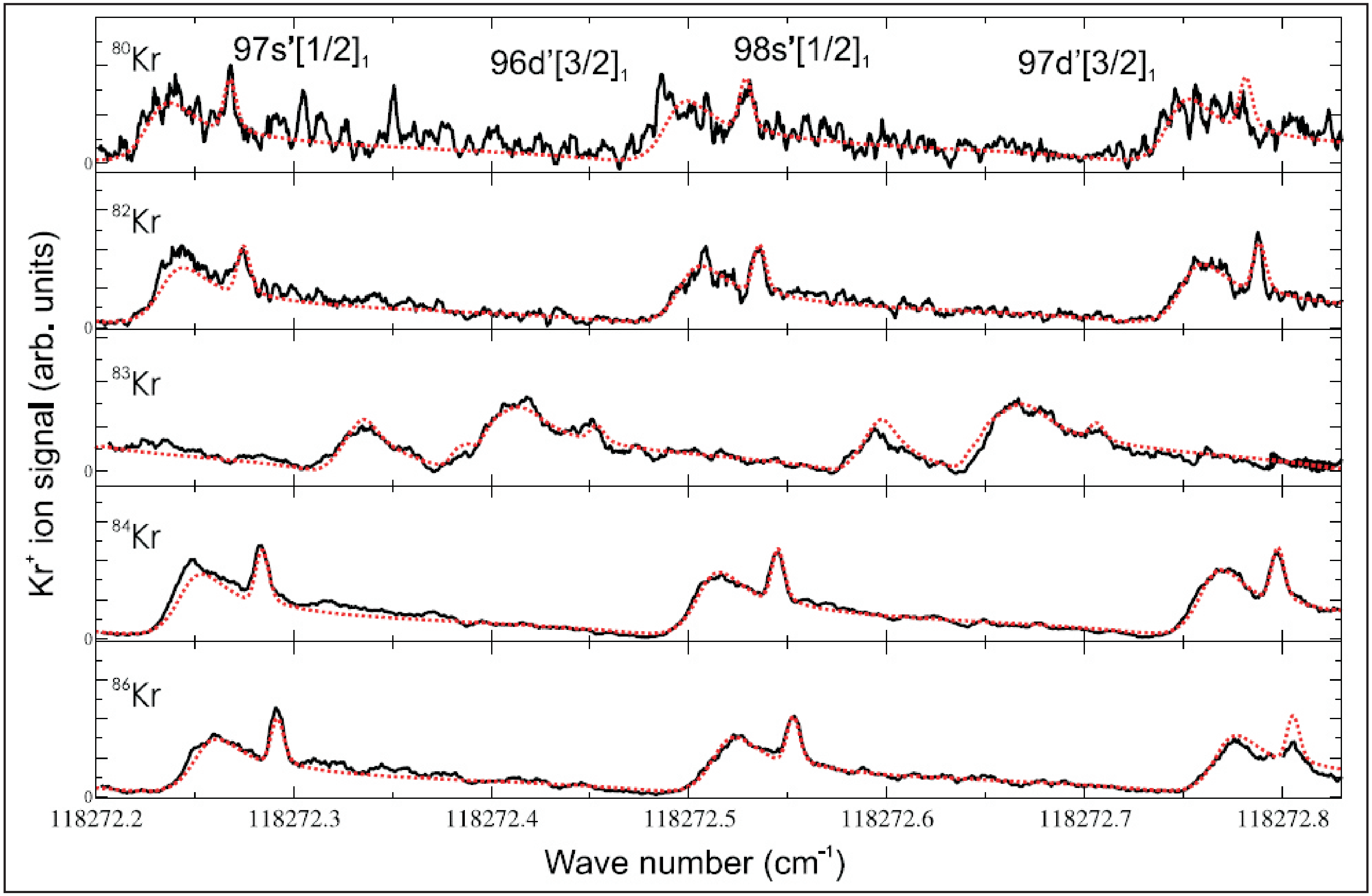
Good quantum numbers: F

core: $S^+=1/2$, $L^+=1$, $J^+=1/2,3/2$, $F^+=3$-6
electron: l=0,2

Quantum defects: must be defined with respect to hyperfine levels of the ion

H.J. Wörner, U. Hollenstein, F. Merkt, Phys. Rev. A 68, 032510 (2003)
see also: J.-Q. Sun, K.T. Lu, JPB, 21, 1957 (1989)

Comparison of MQDT calculations and experiment



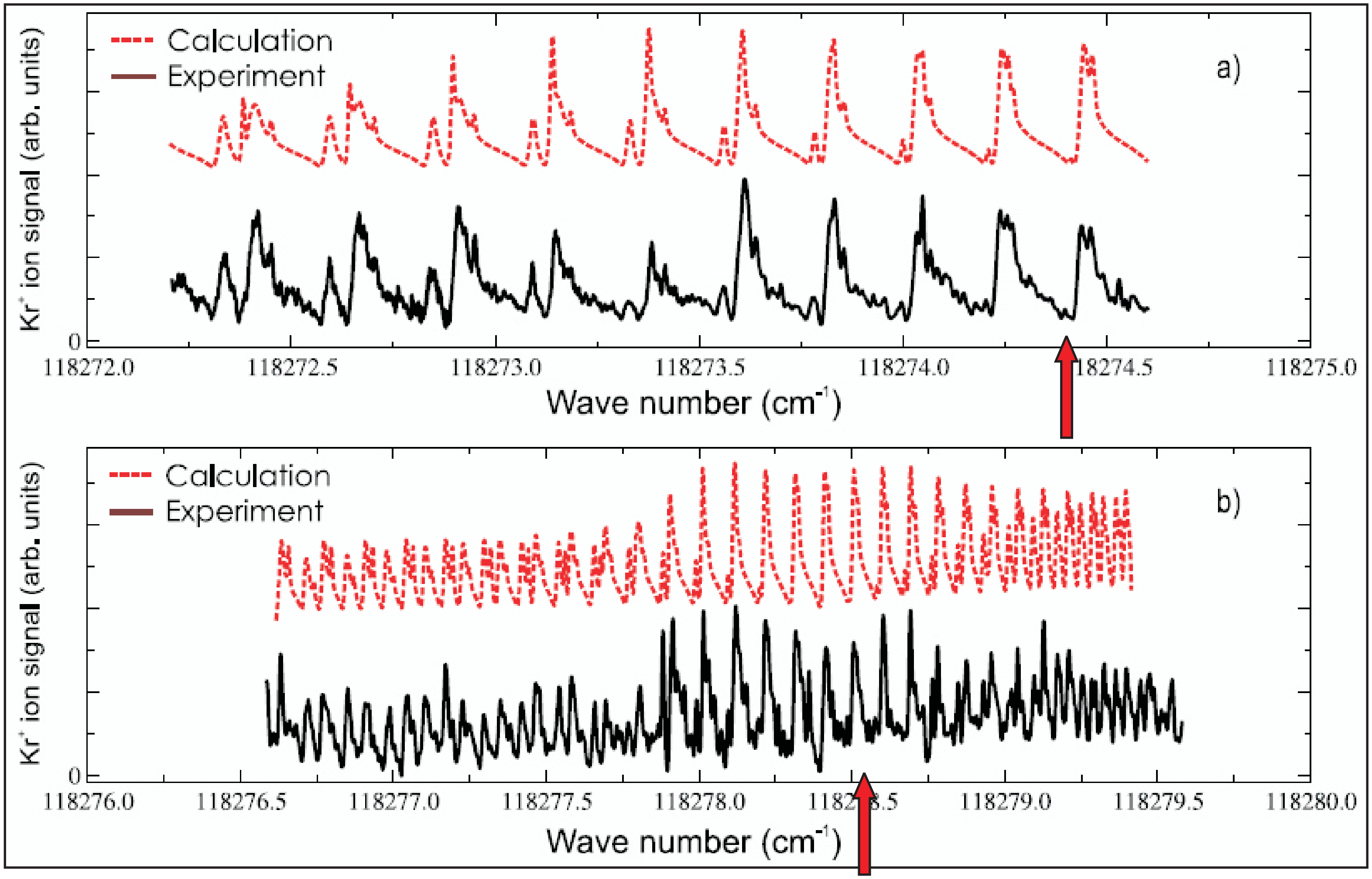
- MQDT parameters: derived in the study of high Rydberg states of Kr below the $^2P_{3/2}$ ionization threshold
- Ionization energies: this work
- Ratio of the transition dipole amplitudes: $D_{d^1P_1} : D_{s^1P_1}$ 1.5 from simulation.

MQDT parameters from: M. Schäfer and F. Merkt. Phys. Rev. A 74, 062506, 2006

Stroboscopic resonances

n = 106, k = 1

n = 133, k = 2



Ionization thresholds

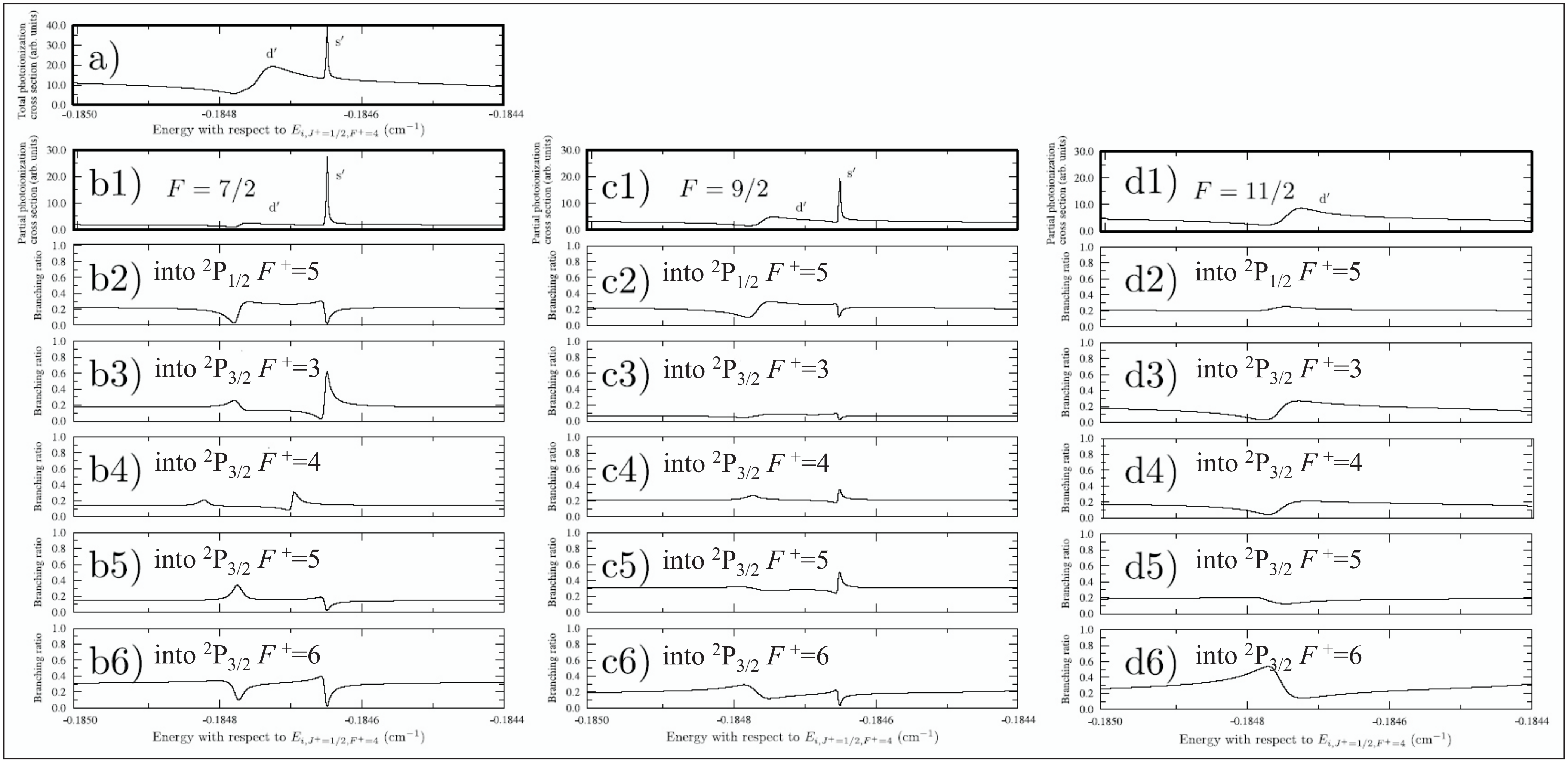
- $A_{1/2}$ of $^{83}Kr^+$: -0.0385(5) cm^{-1}
- Hyperfine splitting of the $^2P_{1/2}$ ionic state: 0.1925(25) cm^{-1},

- Ionization energies and isotope shifts:

Isotope	Natural abundance (%)	Ionization energy $E_{i,J^+=1/2}$ (cm^{-1})	Statistical uncertainty (cm^{-1})	Absolute uncertainty (cm^{-1})	Isotope shift (cm^{-1})
^{78}Kr	0.35(1)	118284.6813[a]	0.0020	0.019	−0.03216(72)[a]
^{80}Kr	2.28(1)	118284.6903	0.0027	0.019	−0.02396(75)[b]
^{82}Kr	11.58(14)	118284.6951	0.0017	0.019	−0.01648(63)[b]
^{83}Kr	11.49(6)	118284.6965	0.0032	0.019	–
^{84}Kr	57.00(4)	118284.7045	0.0017	0.019	−0.00763(57)[b]
^{86}Kr	17.30(22)	118284.7135	0.0019	0.019	0

- The shift per unit of atomic mass: 0.00402(9) cm^{-1}/u
- Average ionization energy: 118284.7036±$(0.0009)_{stat}$ ± $(0.0100)_{abs}$

Partial photoionization cross sections and branching ratios



Conclusions

- A complete survey of isotope effects in the photoionization of Kr has been obtained
  - Average ionization energy: 118284.7036±$(0.0009)_{stat}$ ± $(0.0100)_{abs}$
  - The shift per unit of atomic mass is 0.00402(9) cm^{-1}/u
- Large differences were observed between the photoionization spectra of ^{83}Kr (I = 9/2) and those of the I = 0 isotopes
  - $A_{1/2}$ of $^{83}Kr^{+}$: -0.0385(5) cm^{-1}
  - Stroboscopic resonance
- The spectral patterns were explained by MQDT, which was also used to calculate the branching ratio between hyperfine autoionization and spin-orbit autoionization and different channels.

Acknowledgments:

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